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An examination of the CMAQ simulations of the wet deposition of ammonium from a Bayesian perspective

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Abstract

The ability of the US Environmental Protection Agency's Community Multi-scale Air Quality (CMAQ) model to simulate the wet deposition of ammonium during 8-week winter and summer periods in 2001 is evaluated using observations from the National Acid Deposition Program (NADP) monitoring sites. The objective of this study is to ascertain the effects of precipitation simulations and emissions on CMAQ simulations of deposition. In both seasons, CMAQ tends to underpredict the deposition amounts. Based on the co-located measurements of ammonium wet deposition and precipitation at the NADP sites and on estimated precipitation amounts for each grid cell, Bayesian statistical methods are used to estimate ammonium wet deposition over all grid cells in the study region. To assess the effect of precipitation on the CMAQ simulations, our statistical method is run twice for each time period, using the simulated precipitation information provided to CMAQ and precipitation estimates based on data collected by the cooperative observer network. During the winter period when stratiform-type precipitation dominates, precipitation amounts do not seem to be a major factor in CMAQ's ability to simulate the wet deposition of ammonium. However, during the summer period when precipitation is mainly generated by convective processes, small portions of the region are identified in which problems with precipitation simulations may be adversely affecting CMAQ's estimates.
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1. Introduction

An important aspect in the development and maturation of an air-quality prediction model is the evaluation of the model's ability to predict fields of interest to the air-quality community. This paper focuses on the ability of the US Environmental Protection Agency's Community Multi-scale Air Quality (CMAQ) model to predict the wet deposition of ammonium. A complete description of the

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CMAQ model can be found in Byun and Schere (2006). Two critical elements in this analysis are the precipitation forecasts produced for CMAQ by MM5, a mesoscale meteorological model (Grell et al., 1994; Dudhia et al., 1998), and ammonia emissions. Our objective is to examine the effects of precipitation on the CMAQ simulation of the wet deposition of ammonium. Section 8.2 of Byun and Schere (2006) describes how CMAQ treats wet deposition. There is abundant observed precipitation data with which to judge the MM5 precipitation fields (see the data section). It should be noted that the current implementation of CMAQ does not include scavenging or wet deposition by snow or ice. This would contribute to model uncertainties during the winter season.

The wet deposition of ammonium is an important component of the total mass budget of ammonia/ammonium. Ammonium wet deposition has a detrimental impact on terrestrial and aquatic ecosystems. This is especially true for water quality. For instance, Sheeder et al. (2002) found that nitrate and ammonium were major factors in the decline of water quality in the Chesapeake Bay.

Ammonia emissions are an important factor in understanding and modeling the wet deposition of ammonium. A careful examination of the ammonia emissions data has been made by Gilliland et al. (2003) and Gilliland et al. (2005). Because the main sources of ammonia emissions are fertilizer application and animal husbandry, there is significant uncertainty in the seasonal distribution of the emissions. These two papers and those by Goebes et al. (2003) and Pinder et al. (2004) document the development of an improved ammonia emissions data set. These ammonia emissions data were used in our annual 2001 CMAQ simulation run. The following figures clearly show the problems that CMAQ has in simulating the wet deposition of ammonium. The observed data in these figures came from the NADP monitors. The CMAQ values were obtained at the NADP locations by kriging the CMAQ output fields.

Fig. 1 shows the relationship between the CMAQ simulation of the wet deposition of ammonium and the observed values. Clearly, CMAQ has underpredicted the deposition values in both seasons. The correlation coefficient is particularly low in the summer. We will examine some potential causes for this underprediction. Fig. 2 shows the relationship of the CMAQ deposition simulations to the MM5 precipitation simulations. As one would expect, the deposition values increase as the precipitation increases. The correlation coefficients indicate that the relationship is strong in the summer, but weak in the winter.

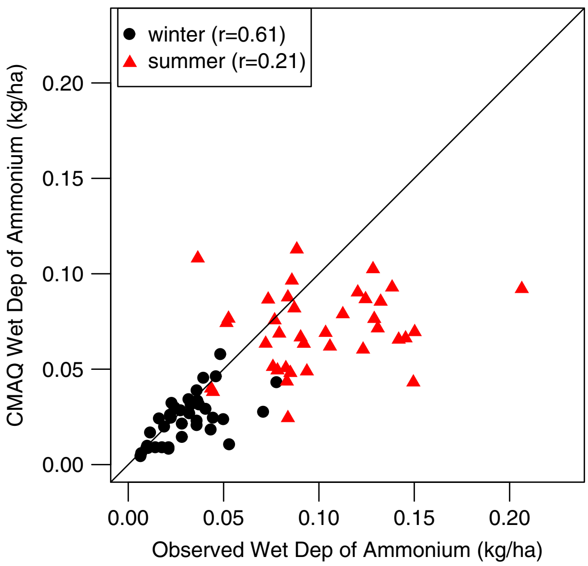

Fig. 1. CMAQ wet deposition of ammonium versus observed wet deposition of ammonium.

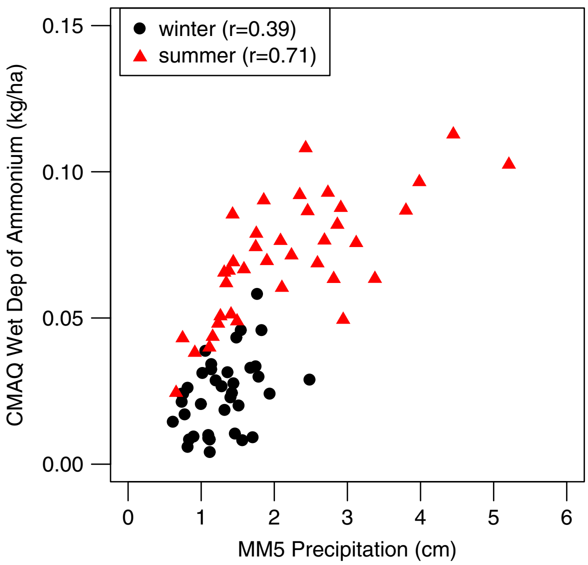

Fig. 2. CMAQ wet deposition of ammonium versus MM5-simulated precipitation.

Fig. 3 shows the relationship between MM5 precipitation simulations and the observed precipitation. The correlation coefficient for the winter is high, while for the summer it is somewhat lower. The convective nature of summer precipitation makes it harder for the model to predict, and this

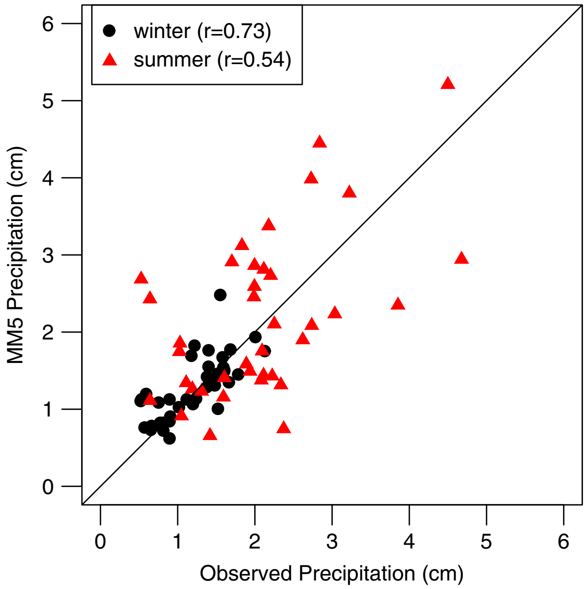

Fig. 3. MM5-simulated precipitation versus observed precipitation.

likely accounts for the lower correlation value. Given the relationship shown in Fig. 3 between MM5-simulated precipitation and the observed precipitation, it seems likely that the CMAQ wet deposition simulations using the MM5 precipitation fields would be comparable to those generated by using the observed precipitation fields. This issue will be examined statistically in this paper.

In this paper, we show how advanced statistical techniques can be used to evaluate the ability of CMAQ to simulate the wet deposition of ammonium, and to assess the impact of MM5-simulated precipitation on these simulations. We evaluate the performance of CMAQ by comparing the simulated values with the observed values of the wet deposition of ammonium. Our approach uses Bayesian statistical methods to explore the nature and the extent of the spatial correlation structure inherent in the observed ammonium wet deposition and to investigate the relationship of observed precipitation and wet deposition. As described by Swall and Davis (2006), the statistical model can then be used to estimate the amount of wet deposition at locations or grid cells for which no monitoring data are available. Major advantages of this particular technique include the ability to quantify the uncertainty inherent in the resulting statistical estimates due to such sources as measurement error, uncertainty about the extent of spatial correlation, and other such stochastic factors. We do not address the statistical aspects of temporal variability.

2. Data

The 2001 calendar year CMAQ simulation was performed on a 36-km horizontal grid using the Lambert conformal projection with 14 vertical layers based on a sigma coordinate system. The simulation used the CB-IV gas-phase chemical mechanism (Gery et al., 1989). The meteorological data were generated by MM5. The MM5 output is processed by the CMAQ Meteorology-Chemistry Interface Processor (MCIP v2.3) to generate input fields to the chemical transport model processor. Emissions data came from the 2001 US Environmental Protection Agency National Emissions Inventory (NEI) for anthropogenic emissions, and from BEIS 3.12 for biogenic emissions (Houyoux, 2004: http://www.epa.gov/air/interstateairquality/pdfs/CAIR_emissions_inventory_overview.pdf).

Our study focuses on an area in the upper midwestern portion of the US, which ranges from eastern North Dakota to western West Virginia; it was selected to encompass a portion of the major source regions, as well as locations downwind from these regions. For reference, this area is shown in Fig. 4. This region of the country is of particular interest due to concerns about possible discrepancies in emissions inventories, mostly attributed to uncertainties about agricultural sources.

Our study focuses on two 8-week time periods. The winter period ranges from 2 January to 27 February 2001, while the summer period includes the weeks from 5 June to 31 July 2001. The observed ammonium wet deposition values and precipitation amounts came from the National Atmospheric Deposition Program (NADP: http://nadp.sws.uiuc.edu) monitoring sites. The NADP data are weekly aggregated samples running from Tuesday morning to Tuesday morning. In our region of interest, there are 50 sparsely distributed NADP sites, a number of which failed to record observations on a regular basis. In addition to the US sites, three Canadian sites which had daily data were also available from the Canadian National Atmospheric Chemistry Database and Analysis System (http://www.msc-smc.ec.gc.ca/natchem/index_e.html).

The observed amounts were averaged over the two 8-week periods. In the winter, only 18 sites had a complete set of data, while 17 sites had one

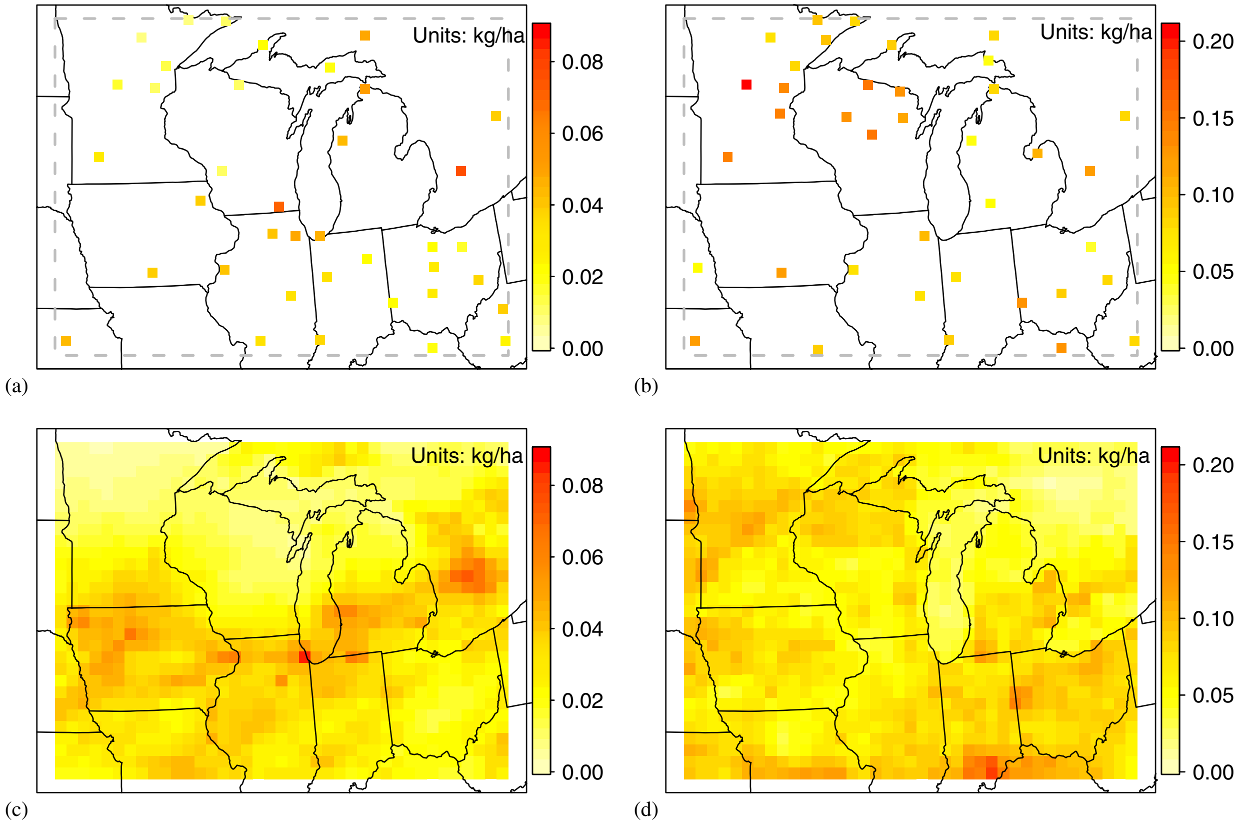

Fig. 4. The top two panels display average weekly NH_4^+ wet deposition measured at NADP sites in (a) winter and (b) summer. For comparison, the bottom two panels display CMAQ-simulated average weekly NH_4^+ wet deposition in (c) winter and (d) summer.

missing week. The remaining 15 sites had multiple weeks of missing data. For the averaging process, we accepted sites that had no more than one missing week, which yielded 35 stations. For the summer period, we had 14 sites with no missing data, and 21 sites with 1 week missing. This again yielded 35 US monitoring sites for use in our analyses. There was minimal missing data from the Canadian locations. Thus, for both summer and winter, a total of 38 monitoring locations were available for this study.

Fig. 4a shows the average weekly ammonium wet deposition amounts observed during the winter period at the NADP monitoring sites within our region of interest, while Fig. 4b shows those available for the summer period. The focus region for the study is outlined in gray on both figures. As demonstrated by the figures and mentioned above, the number of monitoring sites in the vicinity is limited. For comparison, the CMAQ simulated ammonium wet deposition fields were summed for each week, then these weekly deposition amounts were averaged over each of the two 8-week periods. Fig. 4c shows the average weekly values simulated by CMAQ for the winter period, while Fig. 4d illustrates the summer values.

Since our study examines the impact of precipitation errors in CMAQ's simulation of ammonium wet deposition, we now focus on sources of precipitation information in our chosen region. In addition to the observed precipitation at the NADP sites, precipitation observations are available from the US Co-operative Observer Network monitored by the US National Climatic Data Center (www.ncdc.noaa.gov). There are over 1000 cooperative observer sites in the region that were analyzed, each of which measures daily maximum and minimum temperatures in addition to the total daily precipitation. The left panel of Fig. 5 shows the average weekly rainfall used in this study as recorded by the cooperative observers in the region during the 8-week winter timespan, while the right panel shows the amounts recorded in the summer period.

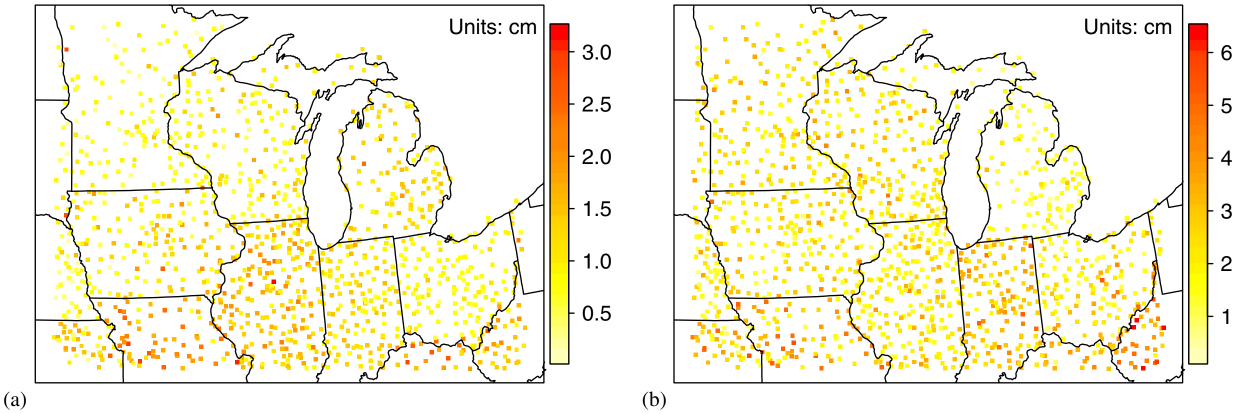

Fig. 5. Precipitation from cooperative observers in (a) winter and (b) summer.

These precipitation amounts are recorded pointwise, with every cooperative observer's location noted. However, we would like to compare and utilize these precipitation amounts with CMAQ and MM5 simulations, which are obtained on a gridded basis. We use a smoothing technique to estimate the amount of precipitation for each grid cell based on the pointwise data, including both the cooperative observers' data and the precipitation recorded at NADP monitoring sites. Any number of smoothing techniques could be used to obtain these values, but we choose to use a simple averaging strategy in which each grid cell's precipitation amount is estimated based on the average of the available monitoring information within a given spatial window. Our averaging window consists of a square region centered at the same point as the grid cell in question, but with a side length of 72 km (as compared with the 36 km side length for a grid cell). Note that this window contains an area equal to that of four grid cells. This allows for more data points to be used in the estimation of each grid cell than if we only considered the monitoring data within each grid cell. Although the cooperative observer network is much denser than that of the NADP, it is still somewhat sparse in certain areas (e.g., northern Minnesota). As a result, there are still some areas in gray on these figures which represent grid cells for which we have no estimate because there were fewer than two observers or monitors within the smoothing window. This includes most portions of the focus region which fall in Ontario, since the cooperative observer network is US-based.

The resulting precipitation estimates for the grid cells in our region in both the winter and summer periods are given in Fig. 6. For reference, the average weekly MM5-simulated precipitation amounts are shown in the bottom panels of the figure. A comparison of the left panels in Fig. 6 shows that areas of low precipitation (in the northwestern and southeastern portions of the focus area) are in agreement during the winter season. However, the band of high precipitation simulated by MM5, which stretched through Missouri and Illinois, is not well established in the observational data. In the summer period (right panels of Fig. 6), MM5-simulated precipitation and the estimates based on cooperative observer data are generally in agreement, although the extremes simulated by MM5 are not very well supported by the observational data. The discrepancies between observed and simulated precipitation amounts are fairly small, and would seem to indicate that errors in precipitation simulations are probably not responsible for discrepancies between observed and CMAQ-simulated ammonium wet deposition. In the next section, we propose a statistical model to help clarify this issue. The paper by Mass et al. (2002) provides a comprehensive review of the ability of MM5 to simulate precipitation under a variety of conditions. Comparisons with radar data are also included as a part of this work as well as the effects of horizontal resolution.

3. Methods

Hierarchical Bayesian analysis provides a framework for the direct assessment of the impact of precipitation on CMAQ simulations of wet deposition. We now outline the theory behind this technique. Our data represent two different levels

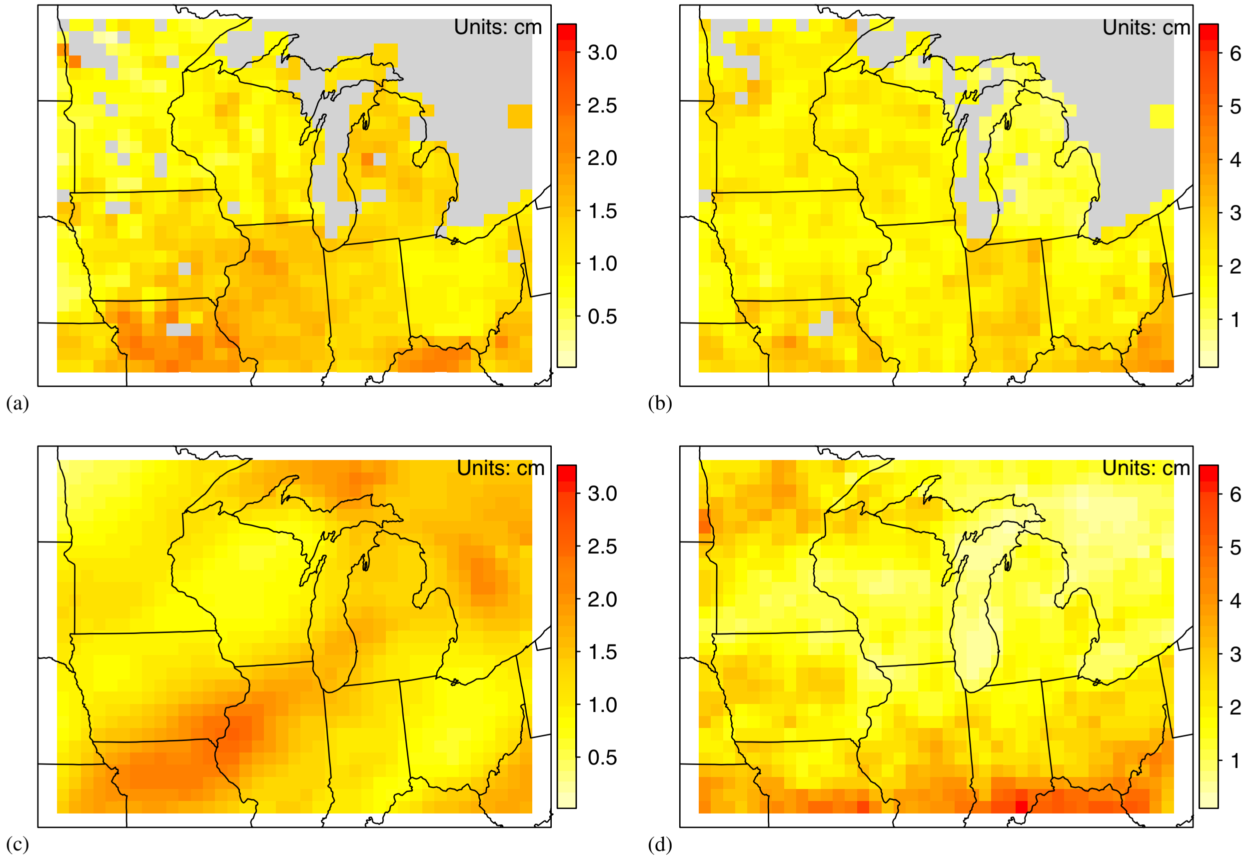

Fig. 6. The top two panels display gridded precipitation from cooperative observers in (a) winter and (b) summer. For comparison, the bottom panels show MM5-simulated precipitation in (c) winter and (d) summer.

of spatial information: pointwise data observed at particular monitoring sites vs. "areal" averages simulated by CMAQ for an entire grid cell. This is an example of the "change of support" problem, as described by Gelfand et al. (2001) and Cressie (1993, Section 5.2). To address this concern, we conduct a further analysis using the Bayesian hierarchical modeling strategy described by Swall and Davis (2006), which was developed based on the work of Fuentes and Raftery (2005). Working on the basis of the (pointwise) observed monitoring data, we use statistical methods to estimate what we would expect CMAQ to simulate for the focus area. We can then compare our statistical estimates for each grid cell with the actual values simulated by CMAQ.

As discussed previously, there is a positive association between precipitation amounts and ammonium wet deposition in both the summer and winter periods. Therefore, CMAQ's ability to simulate wet deposition is at least partly dependent on the quality of information it receives about precipitation, which comes from MM5 through MCIP. This also implies that our statistical model for ammonium wet deposition should include precipitation as an explanatory variable. In order to estimate the average weekly wet deposition amounts for each grid cell over a given time period, we will need an estimate of the average weekly amount of precipitation by grid cell over that period.

We have two possible sources for precipitation information for grid cells. First, we can access the precipitation amounts which CMAQ uses in its simulations; these amounts are simulated by MM5. Fig. 6c displays these values for the winter time period, while Fig. 6d provides those for the summer season. As a second option, we can use the observations taken by the cooperative observers, smoothed to obtain estimated precipitation amounts for the grid cells, as described in the previous section and displayed in the top panels of Fig. 6. If the CMAQ simulations of the wet deposition of ammonium agree more closely with the first set of statistical estimates (based on MM5-simulated precipitation) than with the second set of estimates (based on observational records), then errors in precipitation fields may be the culprit behind inaccuracies in wet deposition simulation. However, we note that since there are some areas where monitoring data are very sparse (represented in gray on the figures), use of these precipitation estimates will prevent us from making statistical estimates for wet deposition in these areas.

Our statistical methodology is described in detail by Swall and Davis (2006), but we summarize the main features of the approach here. For a given 8-week period (e.g., summer or winter time period), we seek to model the average weekly ammonium wet deposition at a given NADP site (denoted a_i) based on the site's location (x_i, y_i), the average weekly precipitation recorded at the site (p_i), and values observed at all the surrounding sites in the region. (Note that the actual CMAQ simulation of ammonium wet deposition does not play a role in fitting this statistical model.) Even after controlling for precipitation, wet deposition amounts are spatially correlated, and this is accounted for in the statistical model using spatially correlated errors. In addition, the model also includes an allowance for measurement error (or other fine-scale error). We combine these assumptions to form a Bayesian hierarchical model for observations from all the sites with the likelihood given by

$$a \sim N(z, \sigma^2 I),$$

where

$$z \sim N(\beta_0 + \beta_1 x + \beta_2 y + \beta_3 xy + \beta_4 p, \Sigma)$$

and I is the identity matrix. The terms β_j denote coefficients describing the association between the location or precipitation for the sites and the observed responses at the sites. As described by Swall and Davis (2006), Σ is a covariance matrix possessing a specified correlation structure; the parameters governing this structure are also estimated by the model. As in Swall and Davis (2006), as well as many other environmental applications, preliminary analyses of the ammonium wet deposition data revealed the exponential covariance structure to be an appropriate choice for the formation of the covariance matrix.

We use Markov Chain Monte-Carlo (MCMC) techniques to sample from the posterior distribution of the statistical parameters; this allows us to assess the combinations of parameters best explaining the relationships among the observational data. To make statistical estimates of the wet deposition of ammonium for each grid cell, we use these combinations of parameters, in conjunction with the locations of the grid cells and the estimates of average weekly precipitation during the time period of interest. This process yields corresponding samples of estimates for each grid cell, from which we can calculate a posterior mean estimate and a credible interval for each. More details about the implementation of the statistical model are provided in the work of Swall and Davis (2006).

As discussed previously, we have two sources of precipitation for grid cells: MM5-simulated precipitation and estimates based on smoothed cooperative observer data. We choose to fit the statistical model twice, once based on each set of precipitation values. As discussed in the previous section and shown in Fig. 6, the MM5-simulated precipitation and the cooperative observer data differ somewhat. One question is whether CMAQ compares more favorably with statistical estimates made based on a particular set of precipitation estimates. For instance, use of an inaccurate precipitation simulation from MM5 (through MCIP) is one factor that could explain errors or biases by CMAQ in predicting wet deposition. If this were a large factor in CMAQ's ability to simulate the wet deposition of ammonium, then the statistical estimates of CMAQ's error in predicting wet deposition of ammonia using MM5-simulated precipitation should be noticeably smaller than those generated using precipitation based on information from cooperative observers. However, if the estimated CMAQ error is similar regardless of the precipitation data estimated for the grid cells, then there are likely other, much more significant causes than errors in the precipitation fields provided to CMAQ. For instance, such factors could include errors in estimating ammonia emissions.

4. Results

We now take a closer look at the estimates provided by the Bayesian hierarchical model discussed in the previous section, beginning with the winter time period. Fig. 7 shows the average ammonium wet deposition estimated by the statistical model using precipitation information from MM5 (left panel) and the cooperative observer

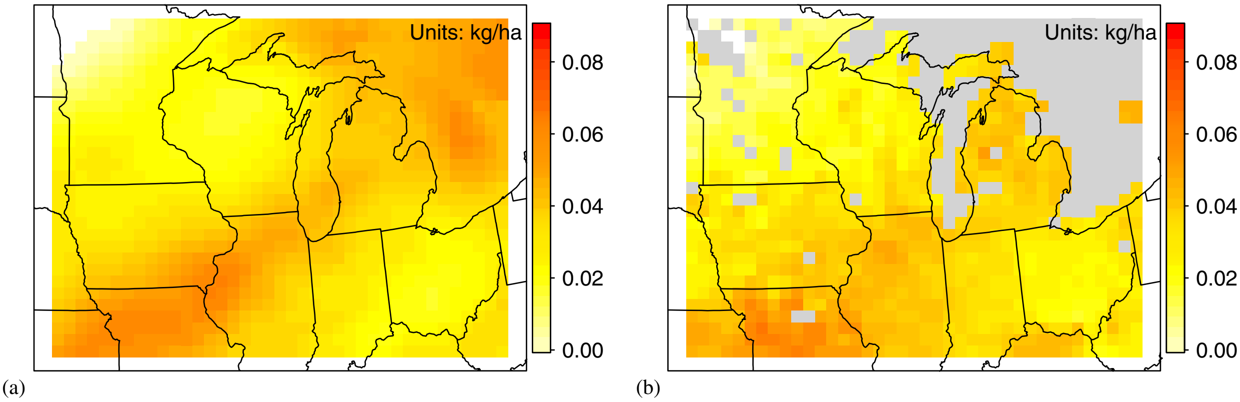

Fig. 7. Winter estimated ammonium wet deposition using (a) MM5-simulated precipitation and (b) cooperative observer precipitation.

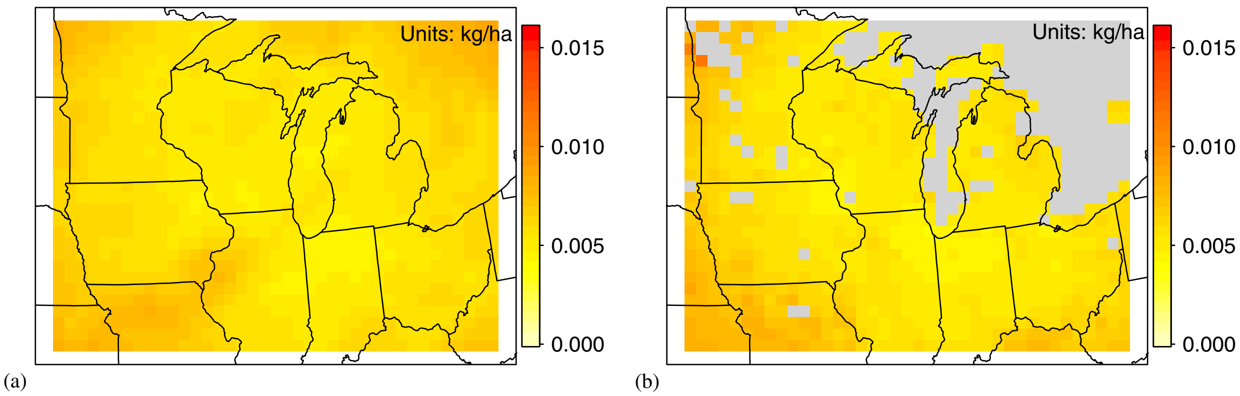

Fig. 8. Standard deviation of winter estimated ammonium wet deposition using (a) MM5-simulated precipitation and (b) cooperative observer precipitation.

network (right panel). A comparison of these two figures reveals some discrepancies, which correspond largely with the discrepancies between the MM5-simulated precipitation (Fig. 6c) and that obtained using the cooperative observer network (Fig. 6a). For instance, this is seen clearly along an imaginary line between northeastern Kansas and northeastern Illinois.

Since these estimates are made on the basis of the Bayesian statistical model described in the previous section, it is fairly easy to take into account the statistical error associated with each estimate and, by extension, associated with the estimated differences between CMAQ and the statistical estimates portrayed in these figures. This variability can be largely captured through the construction of 95% credible intervals, which give the ranges within which the statistical model predicts the ammonium wet deposition levels fall with 95% probability. Then, we can focus our attention on those grid cells in which the simulated ammonium wet deposition given by CMAQ falls outside the credible interval given by the statistical model; these errors are statistically significant. Henceforth, we will refer to the differences (CMAQ-estimated) in these grid cells as significant errors. For reference, Fig. 8 displays the standard deviation associated with the estimated wet deposition for each grid cell, using both MM5-simulated (left panel) and cooperative observer precipitation (right panel). As expected, the uncertainty tends to be higher for grid cells at which there are few or no NADP sites located nearby; for example, variability is noticeably higher in the northwest and northeast corners of the region, where we have no observations. Fig. 9 shows the winter time significant errors, using the

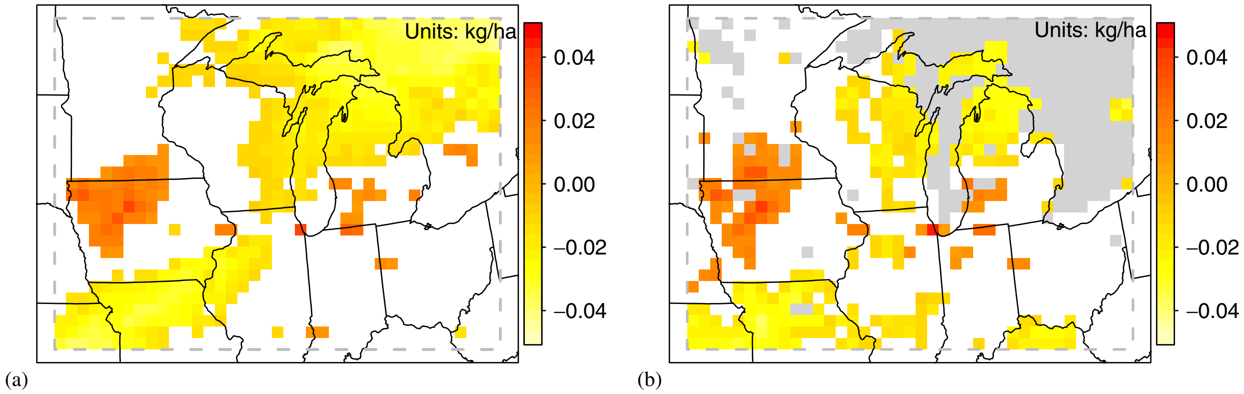

Fig. 9. Winter estimated significant differences (CMAQ-estimated) using (a) MM5-simulated precipitation and (b) cooperative observer precipitation.

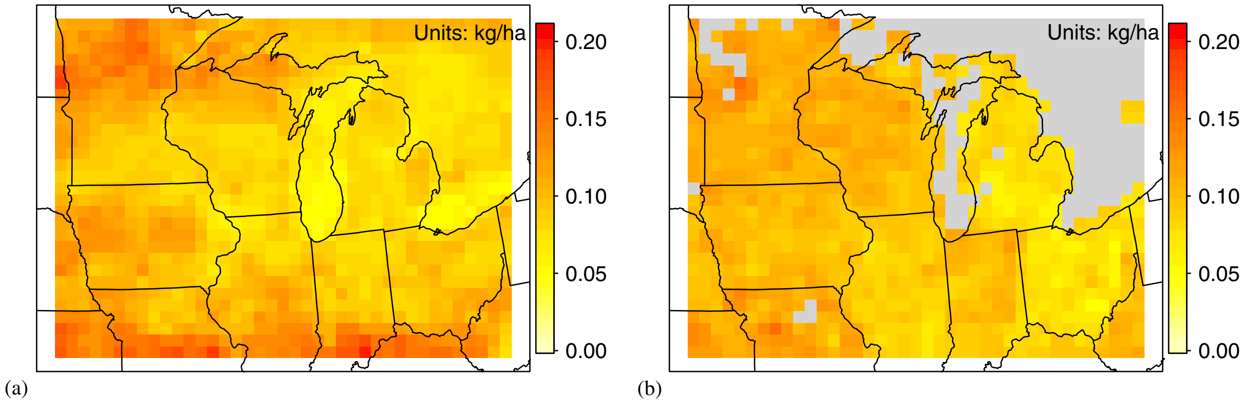

Fig. 10. Summer estimated ammonium wet deposition using (a) MM5-simulated precipitation and (b) cooperative observer precipitation.

MM5-simulated (left panel) and cooperative observer (right panel) precipitation. (Note that non-significant differences are not shown in these figures.) The patterns displayed in these panels are quite similar, even though the precipitation used in the statistical algorithms is from different sources. Both show a substantial overprediction of ammonium wet deposition by CMAQ in the area bridging Minnesota and Iowa and in a limited area in southern Michigan. The areas of underprediction are larger, consisting of much of the southwestern portion of our focus area and portions of eastern Wisconsin and northern Michigan. Of course, without cooperative observers in Ontario, we cannot determine whether the statistical models agree about a suspected large area of underprediction there. One area in which the statistical models disagree is northern Kentucky, where the statistical model based on cooperative observer precipitation suggests that CMAQ may be underpredicting ammonium wet deposition.

Fig. 10 shows the estimated ammonium wet deposition in the summer period using both sets of precipitation data, while Fig. 11 displays the standard deviations of the corresponding wet deposition estimates. Again, as expected, many of the differences we see can be traced back to the differences in the precipitation sources. The most noteworthy of these differences are probably those found along the extreme southern border of the focus region, and in portions of Illinois and Ohio. Interestingly, Fig. 11a shows that for the statistical model which makes use of MM5-simulated precipitation, variability associated with the estimates is highest in this same area, motivating further exploration. We examine further the grid cells in which the CMAQ-simulated ammonium wet deposition differs significantly from that estimated by

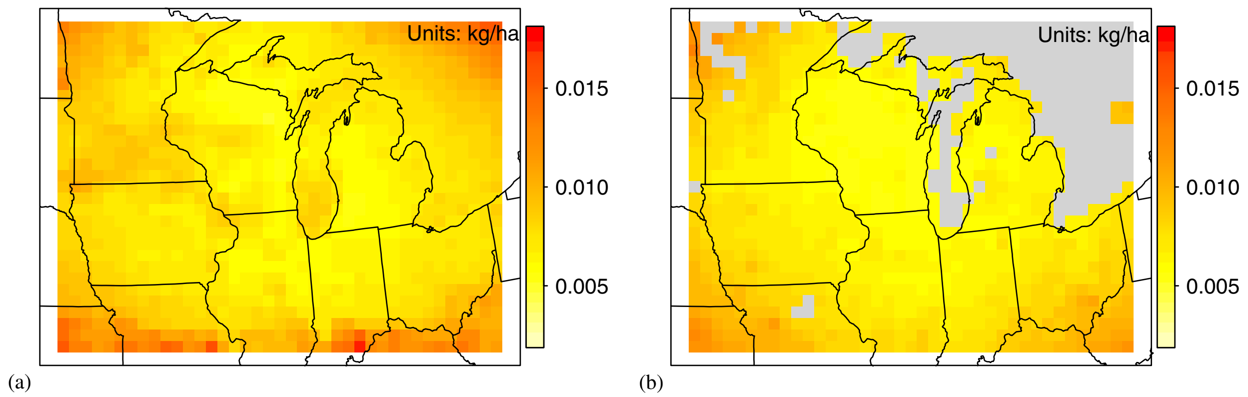

Fig. 11. Standard deviation of summer estimated wet deposition using (a) MM5-simulated precipitation and (b) cooperative observer precipitation.

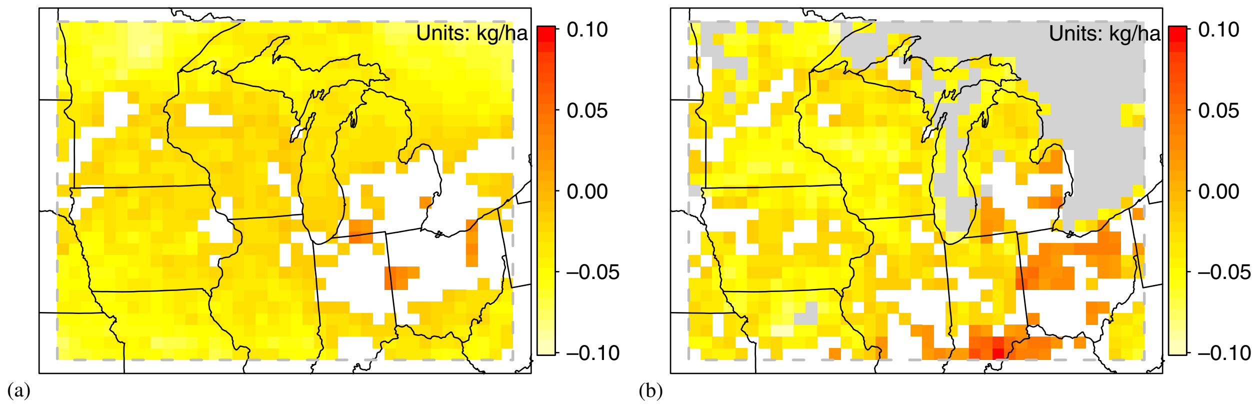

Fig. 12. Summer estimated significant differences (CMAQ-estimated) using (a) MM5-simulated precipitation and (b) cooperative observer data.

the statistical model. Fig. 12 displays these significant errors for the statistical model using simulated precipitation and cooperative observer information in panels (a) and (b), respectively. The western portions of the focus region look similar in both panels, with underprediction of ammonium wet deposition by CMAQ. However, there are several areas where there exists noticeable disagreement. Based on the MM5-simulated precipitation, the statistical model estimates that CMAQ is underpredicting ammonium wet deposition in portions of northern Kentucky and southern Indiana. However, for the same region, but using precipitation data from the cooperative observers, wet deposition is overpredicted in the same area. In addition, there are some areas in central Ohio that are only identified as areas of CMAQ overprediction and areas of Indiana identified as areas of underprediction by the statistical model based on cooperative observer information. These same areas do not appear to have significant errors based on the statistical model using simulated precipitation.

Our examination of the significant differences (winter in Fig. 9 and summer in Fig. 12) has so far been largely an exercise of visual comparison, and we now take a closer, more quantitative approach. For each grid cell, we consider the 95% credible interval for the estimate given by the statistical model using the MM5-simulated precipitation and that using precipitation recorded by the cooperative observers. If, for a given grid cell, the credible interval obtained based on MM5-simulated precipitation and the credible interval obtained using the cooperative observer data do not overlap, then we say that the estimates are significantly different for the specified grid cell. Note that as before, we cannot include grid cells for which we do not have cooperative observer data, so this comparison cannot be conducted in some portions of the focus region. For the summer period, Fig. 13 shows grid

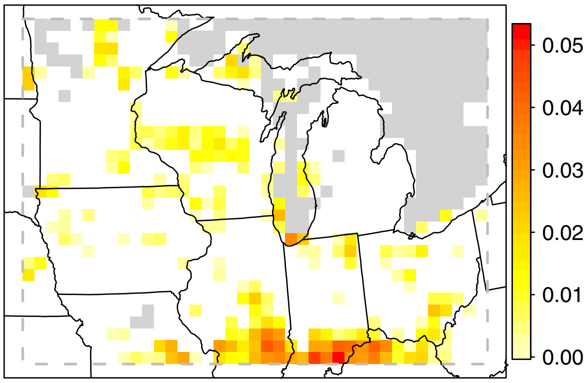

Fig. 13. Absolute differences between credible intervals for grid cells in which statistical wet deposition estimates differ significantly (summer).

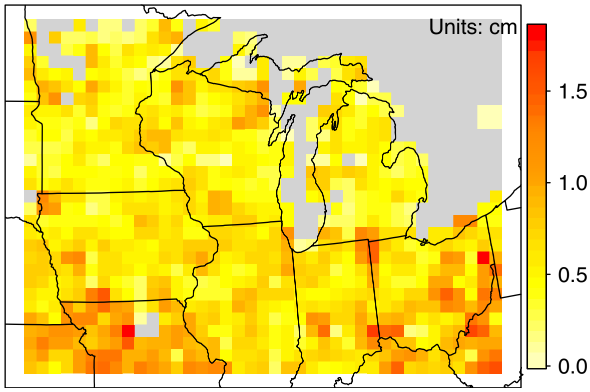

Fig. 14. Standard deviation of precipitation observed within each grid cell (summer).

cells at which the statistical estimates differ significantly. The color assigned to each cell gives the absolute difference between the non-overlapping credible intervals. In addition to problematic areas previously identified, there are also patches of smaller, but still notable, differences in Wisconsin and Minnesota. These areas are worthy of additional investigation to determine any role that precipitation discrepancies may be playing in the CMAQ simulation of wet deposition. During the winter time period, the credible intervals for all available grid cells overlap, so no corresponding figure is included for this time period.

5. Discussion

Comparisons between the significant errors obtained using our two sources of precipitation data are revealing. In the winter period, the pattern of significant errors is very similar, regardless of the source of precipitation data. This implies that although the precipitation data yielded by MM5 and the cooperative observer records are somewhat different, these differences are not a significant factor in the underprediction or overprediction of ammonium deposition by CMAQ. Instead, we might consider other explanations for CMAQ's performance in the winter. One potential issue is the quality of the emissions data, which may be incomplete or may contain poor estimates of ammonium sources, especially agricultural contributions. It is difficult to evaluate the emissions inventory with our statistical algorithm, since we have no observed emission data. However, work has been done in this area using other statistical techniques. See, for example, Gilliland et al. (2003).

In the summer period, the pattern of significant errors differ based on the precipitation information included in the statistical algorithm in a few portions of the focus region. This implies that CMAQ's performance may be at least partially explained by the MM5-simulated precipitation data. During the summer months when convective activity is dominant, we sometimes see a spotty pattern of precipitation that is much harder to model or predict. Monitors located near one another may show differing amounts of rainfall. Fig. 14 shows the standard deviation of precipitation amounts recorded by cooperative observers within the smoothing window for each grid cell in the summer period. As before, grid cells with fewer than two records are colored gray. We note that these standard deviations are high in portions of Ohio and Kentucky, where the patterns of significant errors are different for the two sources of precipitation information, but also high in portions of Missouri and West Virginia.

In areas where rainfall amounts are spatially inhomogeneous, there are two potential problems. The first is that there is more error inherent in the smoothing process, which we use to get precipitation estimates for each grid cell based on the cooperative observers' observations and precipitation information recorded by NADP monitors. Secondly, the statistical model suffers from degraded performance because the ammonium wet deposition field is changing more rapidly in some places than in others; this violates the notion of a stationary covariance structure (Banerjee et al., 2005, Chapter 5), which is inherent in our model. However, as mentioned with regards to the winter period, there is also a considerable amount of uncertainty in the emission inventory, which we cannot assess.

It is clear that there are many additional questions to be addressed. Atmospheric transport processes and their effects on the wet deposition of ammonium have not been addressed in this paper. This is a topic which requires further research. Another approach to the research that we have done would be to take a more limited space/time view to the evaluation of CMAQ; such an approach would be similar to the traditional case-study approach used in meteorology. One could select a limited spatial/temporal domain which possesses precipitation and emissions characteristics that fit a particular research objective. Data-rich regions could then be selected in which to carry out the research, thus putting comparisons between CMAQ simulations and observations on firmer statistical grounds.

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